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Magnetic properties of Ti^{4+} substituted Li–Zn ferrites

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Abstract : Titanium substituted lithium zinc ferrites with the general formula $Li_{0.45+0.5x}Zn_{0.1}Ti_xFe_{2.45-1.5x}O_4$ ($0 \le x \le 1.0$) were prepared by the standard double sintering ceramic technique. Magnetic parameters such as initial permeability (μ_i), saturation magnetization ($4\pi M_s$), coercive force (H_c), retentive force (B_r), remanence ratio (B_r/B_{max}) and Curie temperature (T_c) were measured. A dynamic hysteresis loop tracer was used to measure the hysteresis parameters The values of μ_i, $4\pi M_s$, H_c, B_r and T_c decreased as the content of titanium increased.

Keywords : Li-Zn ferrites, saturation magnetization, Curie temperature

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Lithium ferrites have been extensively studied because of their applications in electronic technology and communication industries. The properties of these materials are sensitive to the type and amount of substitution and preparation methodology. A number of studies [1–7] have been carried out on Li-Zn and Li-Ti ferrites through neutron and X-ray diffraction, magnetization measurements and Mössbauer experiments. Baijal *et al* [8] interpreted the Mössbauer spectra of $Li_{0.45+0.5x}Zn_{0.1}Ti_xFe_{2.45-1.5x}O_4$ at 300 K in terms of the Fe^{3+} ions at tetrahedral (A) and octahedral (B) sites. Electrical properties have been studied [9] for the above ferrites and observed an increase in resistivity with titanium concentration. The present paper reports the modification in the various magnetic properties such as initial permeability, saturation magnetization, coercive force, retentive force, remanence ratio and Curie temperature of these Li-Zn-Ti ferrites with the variation in Ti content. Li-Zn-Ti ferrites with compositional formula $Li_{0.45+0.5x}Zn_{0.1}Ti_xFe_{2.45-1.5x}O_4$ (x varies from 0 to 1.0 in steps of 0.2) were prepared by standard ceramic method using

AR grade Li_2CO_3, ZnO, TiO_2 and Fe_2O_3. Detailed preparation procedure had been given elsewhere [8]. The final samples were in the form of pellets of 10 mm diameter and toroids of 10 mm inner diameter and 16 mm outer diameter and thickness ranging from 3 to 4 mm. The completion of the solid state reaction was confirmed by the X-ray diffraction measurement using Cu-K_α radiation. The saturation magnetization was measured at room temperature using a vibrating sample magnetometer. The Curie temperature measurements were made using a simple method given by Soohoo [10] The initial permeability was determined on the sintered toroids by winding about 60 turns of S.W.G. 30 enamelled copper wire using HP-4275 Å.multi-frequency LCR meter. The hysteresis characteristics were measured with a dynamic hysteresis loop tracer using toroid samples [11].

The single phase formation of the composition of the ferrites has been confirmed from X-ray diffraction pattern. Figure 1 shows X-ray diffraction pattern of some typical

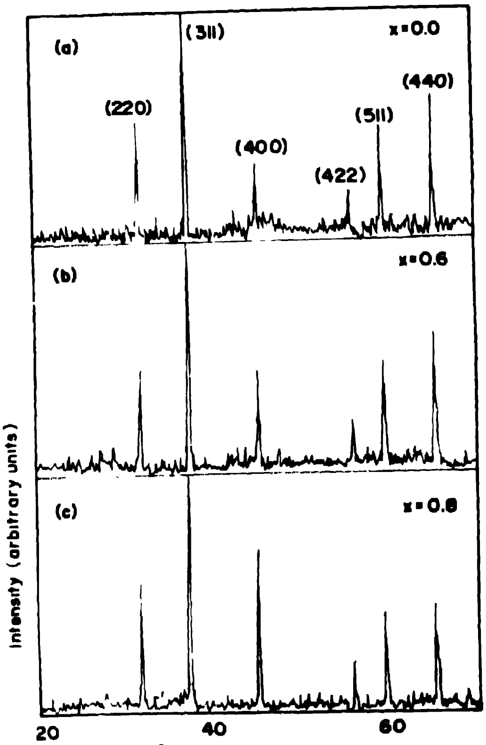

Figure 1. X-ray diffraction patterns of the system $Li_{0.45+0.5x}Zn_{0.1}Ti_xFe_{2.45-1.5x}O_4$ (a) $x = 0.0$, (b) $x = 0.6$ and (c) $x = 0.8$

samples. The variation of the lattice parameter a (Å) with composition is shown in Figure 2 and a slight increase in a was observed. This may be due to the fact that Ti^{4+} ions with larger ionic radii 0.69 Å replaces Fe^{3+} having ionic radii 0.67 Å [1]. Figure 3 shows a decrease in room temperature saturation magnetization ($4\pi M_s$) with progressive substitution of Ti^{4+}. This variation can be understood by considering the cation distribution and the anti-parallel spin alignment on the two sublattice sites following from the Neel's molecular field model [12]. The cationic distribution of these ferrites has been found to be [8].

$$(Zn_{0.1}Li_{0.5x-0.05}Fe_{0.95-0.5x})_A\ [Li_{0.5}Fe_{1.5-x}Ti_x]_B\ O_4 \text{ for } x \geq 0.2. \quad (1)$$

The replacement of Fe^{3+} ions, of magnetic moment $5\mu_B$, on B site by non-magnetic Ti^{4+} ions, leads to a decrease of the magnetic moment of B site (M_B). Substitution with Ti^{4+} also leads to a small replacement of magnetic Fe^{3+} ions at A site by non-magnetic

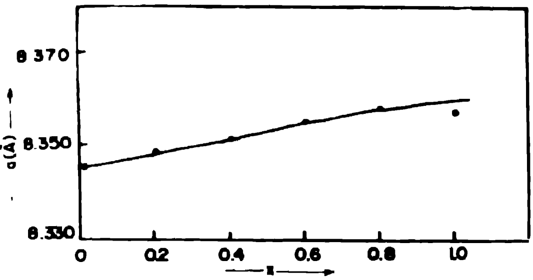

Figure 2. Variation of lattice constant a with titanium concentration x for the ferrite $Li_{0.45+0.5x}Zn_{0.1}Ti_xFe_{2.45-1.5x}O_4$.

Li^+ ions. Hence, the magnetic moment at A site (M_A) is also reduced. However, it is seen from the cationic distribution that the decrease in M_B is slightly more rapid than that of M_A. Therefore, the net saturation magnetization, M_B-M_A, is expected to decrease

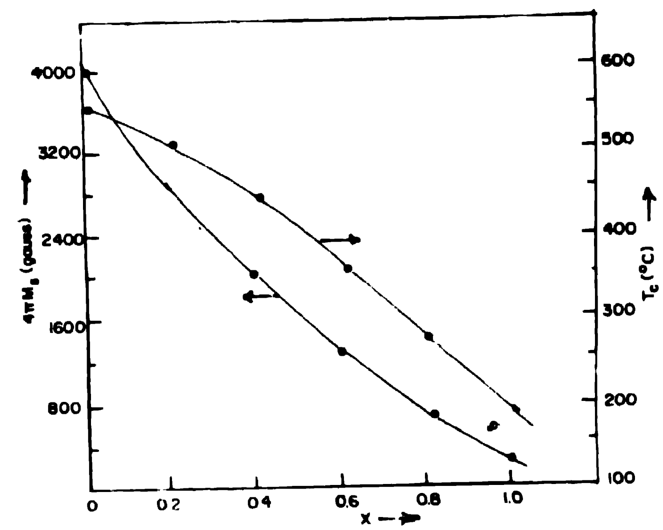

Figure 3. Variation of room temperature saturation magnetization ($4\pi M_s$) and Curie temperature (T_c') of $Li_{0.45+0.5x}Zn_{0.1}Ti_xFe_{2.45-1.5x}O_4$ ferrite as a function of titanium content.

with increase in titanium substitution, as is observed. Theoretically, the influence of Ti^{4+} on the saturation magnetization may be studied by computing the magnetic moment per formula unit from the cation distribution assuming the antiparallel alignment of the

A and B site ions [1,2]. The magnetic moment per formula unit for the above series of ferrites is therefore calculated as

$$n_B = 2.75 - 2.5x \tag{2}$$

and the values are tabulated in Table 1 along with the experimental values. The expected decrease in magnetization with Ti^{4+} is observed in both the experimental and calculated values. However, the slight discrepancy in their values may be due to the fact that the assumed cation distribution as given in eq. (1) may not be exactly the same as the actual cation distribution [1].

Table 1. Magnetic properties of Ti^{4+} substituted lithium-zinc ferrites $Li_{0.45+0.5x}Zn_{0.1}Ti_xFe_{2.45-1.5x}O_4$ $(0 \le x \le 1.0)$

x	Composition	μ_{ic}	n_B Obs	n_B Cal	B_r (gauss)	H_c (gauss)	B_r/B_{max}	T_c (°C)
0	$Li_{0.45}Zn_{0.1}Fe_{2.45}O_4$	62	2.65	2.75	2304	2.24	0.79	575
0.2	$Li_{0.55}Zn_{0.1}Ti_{0.2}Fe_{2.15}O_4$	48	1.88	2.25	1742	1.87	0.80	515
0.4	$Li_{0.65}Zn_{0.1}Ti_{0.4}Fe_{1.85}O_4$	25	1.32	1.75	1332	1.71	0.84	450
0.6	$Li_{0.75}Zn_{0.1}Ti_{0.6}Fe_{1.55}O_4$	21	0.85	1.25	696	1.42	0.75	360
0.8	$Li_{0.85}Zn_{0.1}Ti_{0.8}Fe_{1.25}O_4$	21	0.48	0.75	277	0.79	0.60	280
1.0	$Li_{0.95}Zn_{0.1}Ti_{1.0}Fe_{0.95}O_4$	21	0.25	0.25	137	0.51	0.50	190

The variation of Curie temperature (T_c) is also depicted in Figure 3. According to Neel's model, the AB interaction is most dominant in ferrites. Therefore, the Curie temperature of the ferrites are determined by the overall strength of these AB interaction [13], though sometimes, the intrasublattice AA and BB interaction may become important. This strength is a function of the number of $Fe^{3+}_A - O^{2-} - Fe^{3+}_B$ linkage [14], which in turn depends upon the number of Fe^{3+} ions in the formula unit and also on their distribution on A and B sites. In the present series of ferrites, with the addition of Ti^{4+} ions, the magnetic Fe^{3+} ions are being replaced by non-magnetic Ti^{4+} and Li^+ in the B and A sites respectively. This results in the weakening of AB exchange interactions leading to a decrease of Curie temperature, as has been observed experimentally.

Figure 4 depicts the variation of initial permeability (μ_i) and corrected initial permeability (μ_{ic}) with titanium concentration and a decrease is observed. As the initial permeability of ferrites is greatly influenced by its density, a correction for density is applied according to the relation

$$(\mu_i - 1)_c = (\mu_i - 1)d_x / d, \tag{3}$$

where d_x is the X-ray density and d, the experimental density of the sample. Besides density, the initial permeability is influenced by the ionic distribution, microstructure, composition and method of preparation [15]. According to Went and Wijn [16], the initial permeability is proportional to the square of the saturation magnetization. Hence, the variation of initial permeability with Ti^{4+} concentration follows a similar trend to that of saturation magnetization. According to Brion and Nemback [17], the soluble non-magnetic impurities inside the grain would hinder the reversible motion of domain walls, which can lower the initial permeability. The initial permeability, therefore, decreases with Ti^{4+} substitution. Similar observation has been made by Suryavanshi *et al* [18] in Ti^{4+} substituted Mg–Zn ferrites.

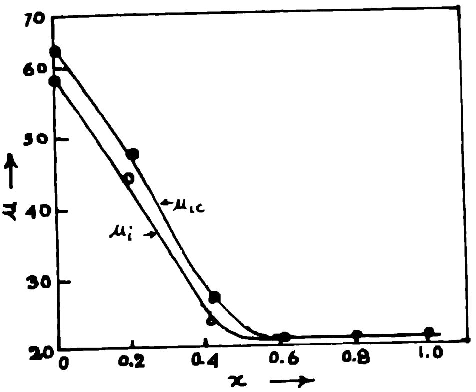

Figure 4. Variation of room temperature initial permeability μ_i (O) and corrected initial permeability μ_{ic} (●) of Li-Zn ferrite as a function of Ti^{4+} substitution.

From the representative hysteresis loop for each of the sample, the magnetic parameters such as retentive force, coercive force, remanence ratio are calculated. Table 1 shows the magnetic parameters of $Li_{0.45+0.5x}Zn_{0.1}Ti_xFe_{2.45-1.5x}O_4$ $(0 \leq x \leq 1.0)$ ferrites. It is observed that the retentive force and coercive force also decreased as the content of titanium increased. However, the remanence ratio is maximum for the sample $x = 0.4$. Thus, it is seen that the substitution of Ti^{4+} in Li–Zn ferrites brings about a decrease in its various magnetic parameters.

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